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An electromagnetic isotope separator

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With 8 figures in the text

Introduction

As is well known, the different isotopes have quite different properties in nuclear physical respects, and therefore it is desirable to produce isotopes in pure form. The electromagnetic method is the most appropriate for this purpose, being the only one that enables complete separation. The first trials with this method were limited by the difficulties encountered in producing suitable ion currents. For the alkaline metals, this is a fairly simple matter; and to begin with, therefore only problems connected with the metals of the first group were studied. Thus, from 1934 to 1938, Li^6 and Li^7 (1, 2, 3, 4) were separated for proton and deuteron reactions, K^{40} (2, 5, 6) and Rb^{87} (7) for investigations of the natural β-activities.

YATES (8) and WALCHER (9, 10, 11) were the first to make use of ion sources of more general types. WALCHER found it possible to make spectroscopic examinations with the separated isotopes of tallium and rubidium (9, 11).

KOCH (12, 13) investigated the possibilities of separating and collecting such substances which exist in gaseous form at atmospheric pressure. BROSTRÖM, HUUS and KOCH (14) determined the proton resonances in separated neon isotopes. In 1940, a number of uranium separations were made by spectrometers for the study of fission (15, 16, 17, 18). In some cases, it has been found possible, with precision spectrographs, to identify artificially radioactive isotopes (19, 20, 21). Nevertheless, when it is desired to determine both the mass number and the half life period of a separated isotope, it is necessary, in most cases, to use an isotope separator for high ion currents.

The isotope separator to be described here has been designed to be able to separate isotopes of all the elements within reasonable times and in such quantities as most nuclear physical experiments would require. These amounts vary from fractions of μg (1, 16) to several mg. Thus for proton resonance determinations it is desirable to work with very thin layers, down to some $\mu g/cm^2$ (22, 14) in order to keep the line-width down. For certain extreme cases however even greater amounts may naturally be considered desirable. Plants of considerable dimensions, such as the calutron and similar types used in U. S. A. during the war, are then required.

Table 1 shows the ion currents required to produce 10 μg hour.

Table 1

Mass number	Ion current (μA)
10	26.8
30	8.9
60	4.5
100	2.7
150	1.8
200	1.3
235	1.1

General requirements on the isotope separator

In the choice of focussing principle there are mainly two possibilities to be considered. Either the ion beam produced in the ion source and then accelerated may be allowed to emerge divergently, in which case the 180° focussing method has to be used, or the electrostatic focussing system may be arranged in such a way that the ion beam emerging therefrom becomes parallel, in which case the 90° focussing method may be applied. The latter of these principles has been used in the separator described here (see Fig. 1).

The following qualities were required of the isotope separator:

1) The ion source should be capable of effectively producing ions of gases and metals.

2) The production of ions should proceed at a low pressure and the pumping speed be so great that no gas scattering occurs in the space, where the ions have low energy.

3) The ion source must not give rise to energy spread in the ion beam.

4) The drawing out of ions from the ion source should take place without ion current losses to the lens system.

5) The diameter of the lens system should be so large that spherical aberration is small even for the beam diameters required with high ion currents, to diminish the effects of space charge.

6) The magnetic fields and the acceleration voltage must be carefully stabilized.

7) The magnetic field should be homogeneous and easily variable.

8) The isotopes of the heaviest elements with a mass difference of 1 should be completely resolved.

The ion source

Arc ion source of probe type has been avoided since there was reason to suppose that the energy dispersion would be of the magnitude of the probe voltage (4). An ion source of Zinn type was tried as diffusion source and was found to give total current intensities up to 1 mA with argon. It works however with a high pressure, requiring differential pumping, which is technically a complication. An ion source of magnet type (see Fig. 2) proved to be the most convenient for our purposes. It

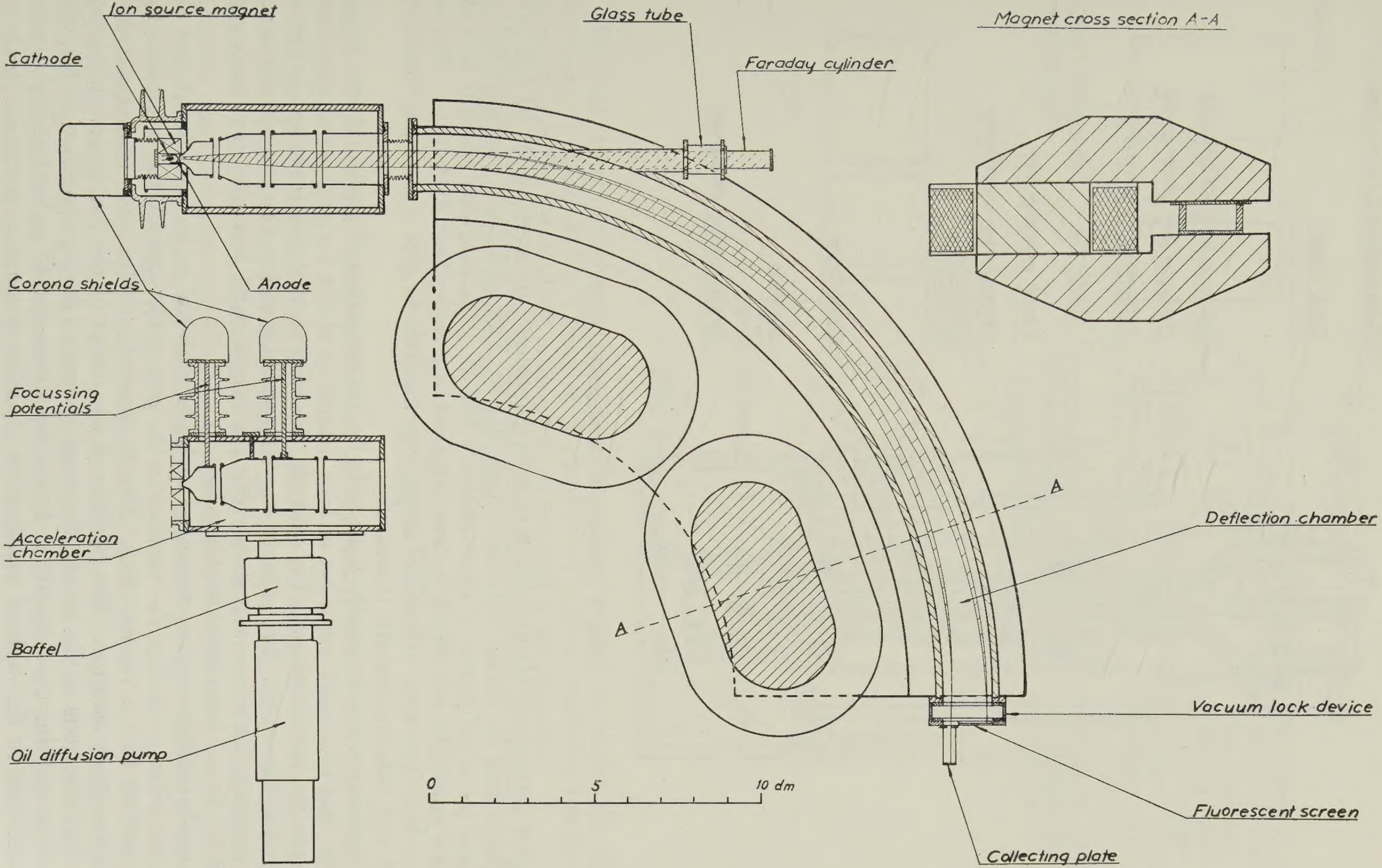

Fig. 1. Cross-section of the isotope separator.

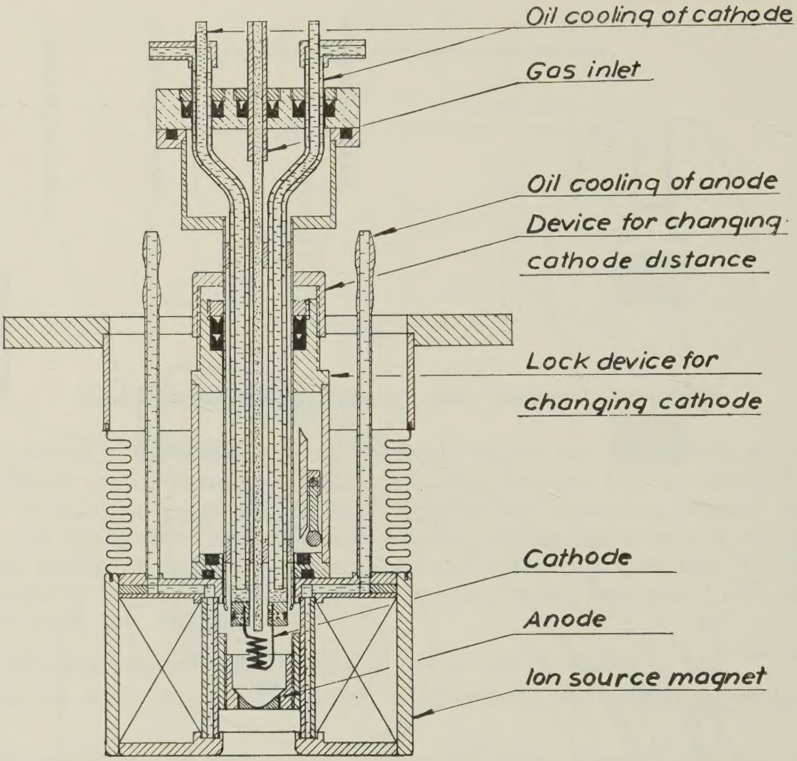

Fig. 2. Ion source of magnetic type.

works at a pressure of about 0.001 mm with continuous arc currents up to 2 A. The arc voltage may be varied between 0 and 200 V. The maximum magnetic field is 600 Gauss for a coil power of approximately 150 W. The tungsten cathode and the anode are cooled by oil, which in turn is cooled in a water heat exchanger. One ion source of this type used to produce argon ions was able to give a total ion current of max. 2 mA calorimetrically measured.

The ion source was originally built only for investigation of gases. It was found, however, that if a small container of lava loaded by about 1 g of a metal was introduced into the ion source near the cathode, the heat from the cathode was sufficient to evaporate the metal. The ratio of the evaporated metal to the amount of the collected isotopes for silver was about 10^4. For longer runs, however, it is more convenient to use a separately heated oven.

To enable cathodes to be exchanged and new material to be introduced into the ion source without spoiling the vacuum in the rest of the system, the ion source has been provided with a vacuum lock. This is especially useful when separating short lived radioactive isotopes. The position of the outlet of the ion source in relation to the first electrode of the lens system may be varied during operation within wide limits, thus ensuring good centring. As the ion source should be kept at high tension it is connected to the earthed acceleration chamber through a porcelain insulator.

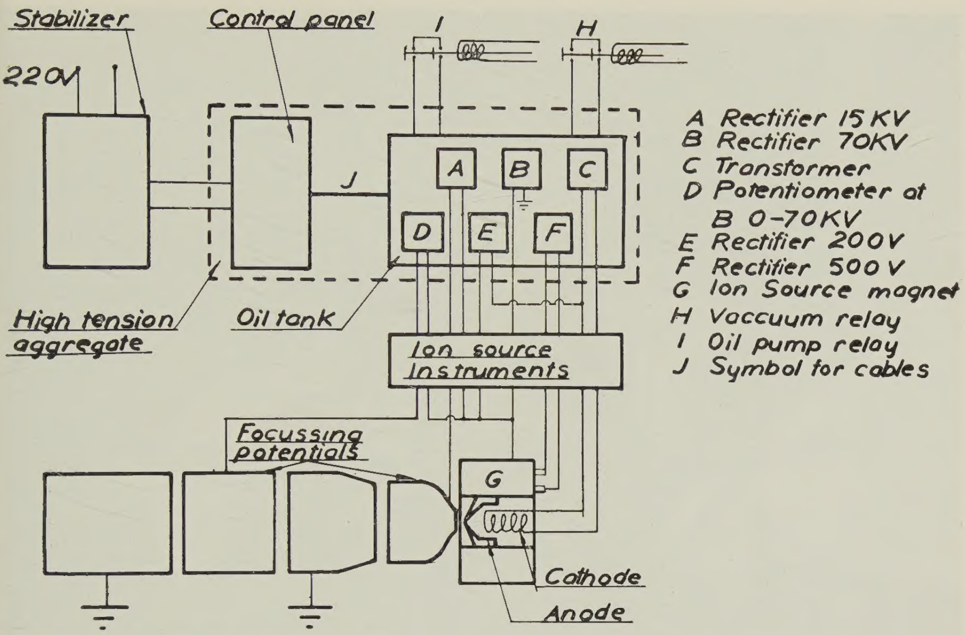

Fig. 3. Block diagram of the high voltage supply.

The lens system

The lens system consists of four chromium plated copper electrodes, the first three of which are suspended in a large brass chamber (Fig. 4) so that they can be centred in relation to the third which is attached to one end of the chamber.

When the beam diameter in the deflection chamber is 65 mm the line broadening on the target for this reason will be about

$$b_d = \frac{d^2}{8\,r} = \frac{6.5^2}{8 \cdot 160} = 0.033 \text{ cm}$$

where, d = the ion beam's diameter at the entrance to the deflection tank and r = magnet's radius.

The electrode diameter for the two last lenses was made 150 mm, as spherical aberration increases rapidly when the beam diameter exceeds about 0.4 of the lens diameter. The electrode diameters at the first lens is 100 and 110 mm. The distances between the electrodes are 15 mm. Observation windows have been located right in front of the lens gaps, in the acceleration chamber, thus making it possible to follow the beam visually (Fig. 4).

The potential of the first electrode can be varied between 0 and 15 kV with respect to the high tension V_B (max. 70 kV). The second and the fourth electrodes are earthed, while the potential V_D of the third electrode should be about 60% of V_B for parallel focussing of the beam. The three last electrodes which form a unipotential symmetrical lens, would by itself be sufficient for parallel focussing, (12). With that system

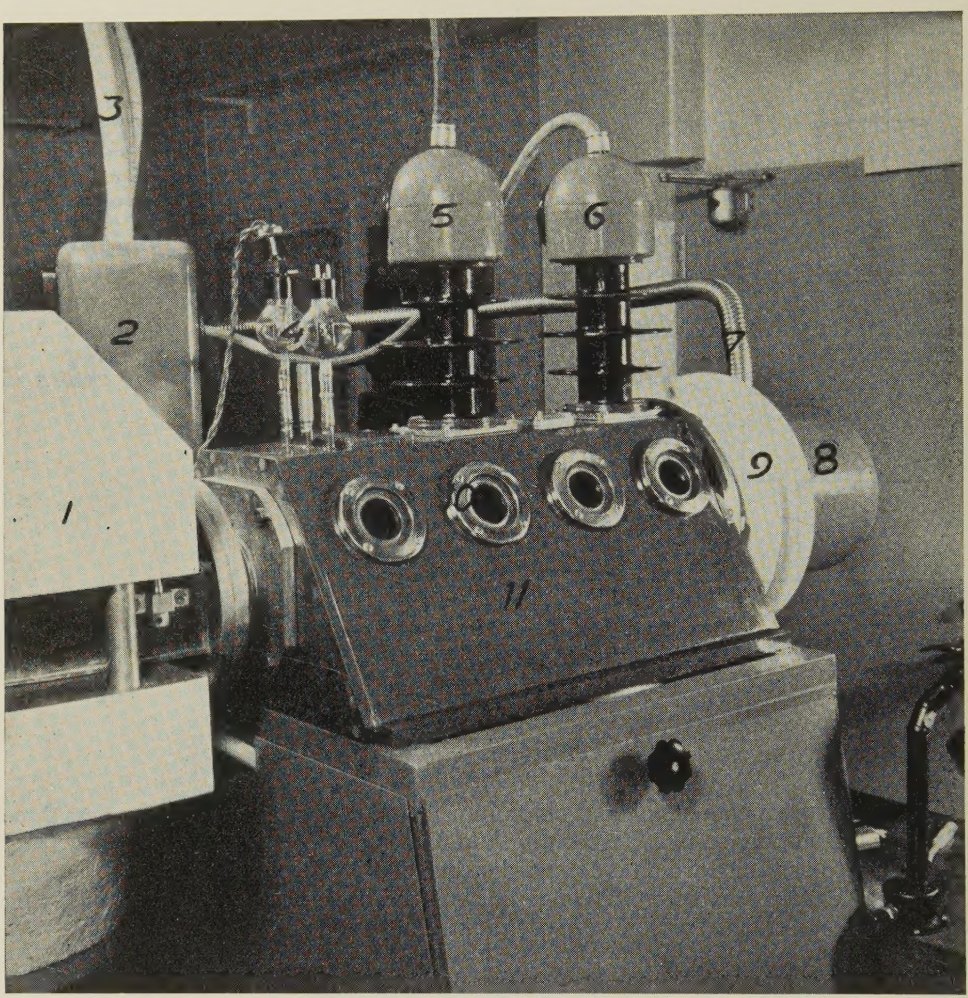

Fig. 4. Accleration chamber.

1) Magnet.
2) Ion source instruments.
3) Cables from 18 (fig. 5).
4) Ionization gauge.
5) Corona shield for connection of third electrode.
6) Corona shield for connection of first electrode.
7) Cables for ion source voltages.
8) Corona shield for ion source.
9) Ion source insulator (70 kV).
10) Observation windows.
11) Acceleration chamber.

alone, however, it would not be possible to vary the beam width at will, which is desirable for strong ion currents in order to bring down the space charge effects to a minimum and to be able to obtain suitable line shape for irradiation experiments.

The ion beam emerges from the smaller hole in the first electrode with a certain opening angle, which decreases for increased potential difference V_A between the ion source and the first electrode. At the same time, however, the refracting power in

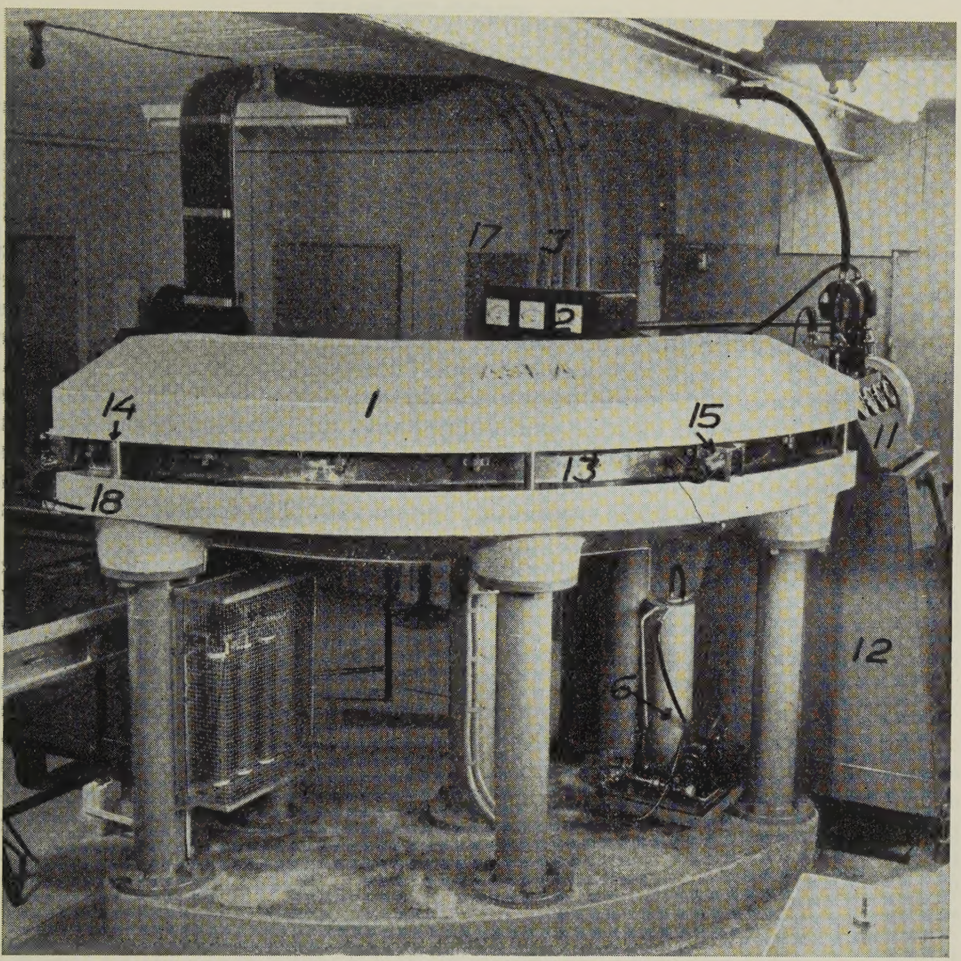

Fig. 5. View of the separator.

1), 2), 3), 11) see fig. 4.
13) Deflection chamber.
14) Brass spacer.
15) Faraday cylinder for measuring the undeflected beam.
16) Oil pump for cooling the ion source.
17) Stabilizer for the magnet current.
18) Control of potentials for the ion source and electrodes.

the first lens decreases. The latter effect is greater than the former and the final result is that the diameter of the ion-beam in lens gap 2 and therefore in the final parallel focussed beam increases as V_A increases. Large changes in V_A require only small changes in V_D for parallel focussing.

All tensions for ion source and electrodes are generated in small oil insulated units mounted together to a single unit (see Fig. 3). For a total current of 1 mA the acceleration voltage ripple will be < 25 Volts. The line width thus arising will therefore be given by

$$b_E = \frac{r}{2}\,\frac{\Delta E}{E} = \frac{160}{2}\cdot\frac{25}{70000} \approx 0.028 \text{ cm.}$$

The primary tension is stabilized for voltage fluctuations and particularly for slow frequency changes by a Sörensen regulator.

The magnet and its equipments

The radius of the magnet is 160 cm, distance between pole pieces (Fig. 1) 10.3 cm and their width 35 cm. At a distance of 7.5 cm on either side of the centre line the magnetic field deviates $< 1‰$ from the value at the centre line. The pole pieces are adjusted mechanically to an accuracy of 0.02–0.2 mm. The total weight of the magnet is 6.5 tons. To prevent changes in pole distance at different magnetic fields, brass spacers have been placed outside the deflection chamber. The chamber is made of copper, screwed together and soldered with tin. The top of the chamber is shaped to a lid and tightened by a packing about 5 m long. To be able to measure and observe the non deflected beam, the chamber has been provided with a side tube in the prolongation of the acceleration system axis. The acceleration chamber is connected to the magnet chamber by a metal bellows.

The magnetic field is produced by two water cooled coils. When the magnetic field is $\approx$ 3 500 G and the acceleration voltage is 60 kV, the heaviest elements can be separated. The current in the coils is then $\approx$ 164 A and is delivered by a 9 kW motor generator. The requisite power for this magnetic field is 7.8 kW.

The magnetic field is stabilized (Fig. 6) by a high speed regulator (ASEA type YRLBE). A similar equipment has for many years been used for the Institute's smaller cyclotron (23), which is only voltage stabilized, however. A change of resistance in the coils of only $1‰$ gives rise to a line drift of 1.6 mm. In order to prevent a drift of the isotope spectrum during the heating of the coils a current stabilization is indispensible. The stabilization accuracy is for the lower mass numbers around $\pm 0.6‰$ and, for the higher mass numbers around $\pm 0.3‰$. The frequency of the changes is low, and for the higher mass numbers the amplitude in such a shift will be < 0.5 mm.

The voltage regulating device in the stabilizator of the regulator N is the coil s (see Fig. 6), which receives a part of the entire magnet voltage through the potentiometer F. Any change in the voltage over s changes the time during which c remains in contact and thus also the impulses, issuing from bridge H and actuating the booster generator D, which in turn alters the magnetizing current of the main generator D. The arm vibrator a is maintained in oscillation by the impulse transformer G.

The total magnetic current gives a voltage drop over the shunt K which controls the compensation amplifier L. Changes in the magnetic current give rise to impulses to a relay T which increases or decreases a motor operated part of the potentiometer F. Thus also current variations affect the voltage regulating coil s.

Collector device

The isotope spectrum is observed visually on a fluorescent screen of zinc silicate. The collection of the isotopes can be done on a metal plate, e. g., Cu Al W Ag and so on, dependent on the investigation it is intended to use the isotopes for. During the adjustments of the focussing the position of the plate is flat to the floor of the

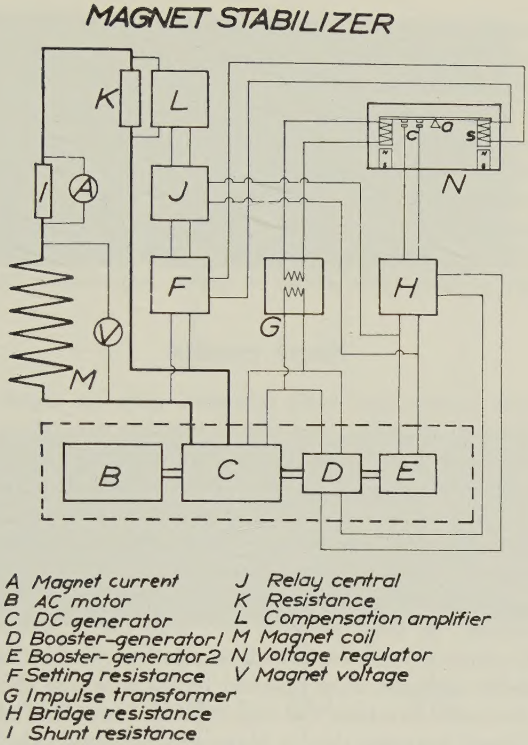

Fig. 6. Block diagram of the magnet stabilizer.

deflection chamber so that it does not obstruct the ion beam. It is connected to an insulated shaft entering the vacuum lock chamber through a vacuum junction and during collection it can be set vertical to intercept the ion beam. The ion current collected by the plate is electrically measured throughout the separation time without Faraday device, as the collector is in the strong stray field of the magnet. To enable the target rapidly to be changed when separating short lived active isotopes, the device is furnished with a vacuum lock.

Pump system

The rough vacuum pump employed is a Kinney pump (pumping speed 3.3 lit/s at 0.01 mm). In order to arrive more rapidly at good vacuum, one of the Institute's disk molecular pumps (pumping speed 160 lit/s) was coupled in between the rough vacuum pump and the oil diffusion pump (500 lit/s at 10^{-4} mm). The diffusion pump is connected to the bottom of the acceleration chamber. The pressure in the system is $\approx 2\times10^{-5}$ mm during the separation time.

Control system

All the instruments and operating devices have been grouped around the collector, so that the whole plant can be operated by one man. Thus the switches for the pumps and the magnet current, the regulating devices for the magnet current, the detection instruments the ionization gauge and the Pirani gauges have been located on a common panel. The upper part of the high tension unit is shaped to form a control board with primary regulation of all partial voltages to ion source and lens system. The partial voltages are conveyed via a high tension insulated instrument cabinet to the connections for electrodes and ion source respectively (Fig. 5).

A relay system ensures that the heater current of the diffusion pump, the magnet current or the filament current of the ion source is broken as soon as the respective cooling systems fail to operate. A special relay breaks the high tension when the pressure in the vacuum system rises above a given adjustable value, thus avoiding discharges.

Some results

After the focussing system has been trimmed with an argon ion beam the first separation was performed with Hg, in order to test the resolution in a case where the isotopes lie particularly close together. The problem was also to decide which of the isotopes Hg^{203} or Hg^{205} is β-active with the half life of 53 days. A pile sample from Oak Ridge, the radioactive radiation of which has been further studied by H. SLÄTIS and K. SIEGBAHN (24), was labelled $Hg^{203, 205}$. A very small quantity of the sample ($HgNO_3$) was transformed to metallic Hg in a glass tube by evaporation. By means of a heated spiral around the glass tube the Hg was continuously evaporated into the ion source. A total amount of 7.5 μg for all the lines was collected in about 45 min. with about 0.5 μA for the strongest line. Collection was done on a Cu plate, where the stable isotopes were marked by light spots, thus making it easy to fix the position of the mass numbers 203 and 205. A 1 mm lead absorber with a 0.5 × ×20 mm slit was placed between the Cu plate and a β tube. The Cu plate was moved 0.25 mm by a micrometer screw after each activity measurement. Fig. 7 shows the activity as a function of the position of the micrometer screw (in mm). The positions of the mass numbers have been measured and drawn in on the figure and from this it is seen that Hg^{203} is the active isotope, which agrees with the suggestion of INGHRAM, HESS and HAYDEN (20). In this separation the focussing of the beam was not

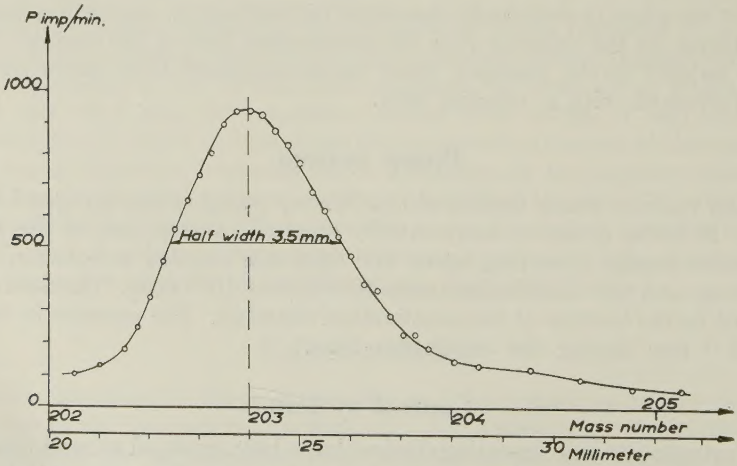

Fig. 7. Mass number determination of the 53^{d} mercury isotope.

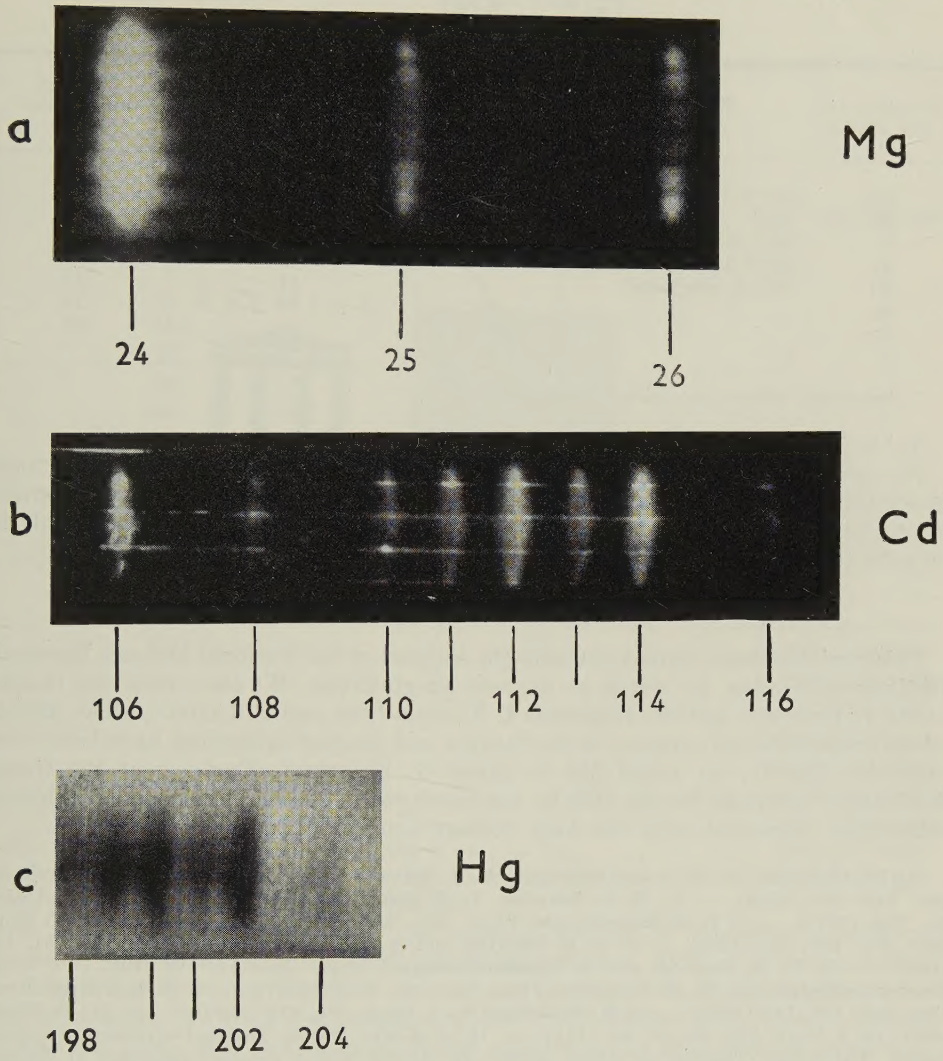

Fig. 8. a and b: Photo of the Mg and Cd spectra as seen on the fluorescent screen.
c: Mercury spectrum deposited on an aluminium plate.
All spectra are photographed to scale. The line distances agree very well with the calculated ones.

so good and the line width was rather large. Fig. 8 c shows Hg spectrum separated on an Al plate when better focussing was attained. Fig. 8 a and b shows the Mg and Cd spectrum photographed on the fluorescent screen. The apparent brightness of the mass number 106 is due to an over-sensitivity of this part of the screen.

Similar measurements have been carried out with Ag, bombarded with deuterons in the Institute's 6.5 MeV cyclotron (32″). In the same way as Hg it was possible to confirm that the 225 *d* activity belongs to Ag^{110} and the 13 h activity in palladium belongs to Pd^{109}.

Table 2

Substance in ion source	Separated isotopes	Amount collected (μg)	Highest current intensity during sep. (μA)	Time of separation (min)
Li*	Li^6 Li^7	50	250	200
Mg	Mg^{24}	50	100	270
Se	Se^{78} Se^{80}	70	25	150
Ag	Ag^{107} Ag^{109}	150	50	52
Cd	Whole spectrum	8	11	15
Sn	»	38	50	18
Hg	»	100	15	60
U	U^+	—	8	—

* Separated with a provisional arrangement.

Table 2 shows the values obtained in the first metal separations.

No great efforts have been made in these first separations to use the most favourable conditions for maximum ion current. Nevertheless, the trials carried out so far definitely show that with the present apparatus it should be possible to increase the yields substancially since higher ion currents can be obtained.

Acknowledgements

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